EXECUTIVE SUMMARY

I. INTRODUCTION

This report has been prepared to summarize the findings of the Oak Ridge Gaseous Diffusion Plant (ORGDP)[1] Mass Balance Project and to support preparation of associated U. S. Department of Energy (DOE) site reports. The project was conducted to support DOE efforts to assess the potential for health and environmental issues resulting from the presence of transuranic (TRU) elements and fission products in recycled uranium (RU) from reactor returns that was processed by DOE and its predecessor agencies. The U.S. Government used uranium in fission reactors to produce plutonium and tritium for nuclear weapons production. Because uranium was scarce relative to demand when these operations began almost 50 years ago, the spent fuel from U.S. fission reactors was processed to recover uranium for recycling. DOE's reconstruction of the historical flow and processing of RU has three fundamental elements:

- Determining annual mass flow of RU throughout the DOE system from the start of processing to March 31, 1999.
- Identifying the characteristics and contaminants (e.g., Pu, Np, and ^{99}Tc) in the major uranium streams.
- Conducting at appropriate sites mass balance activities sufficient to identify any significant implications for personnel exposure or environmental contamination.

The ORGDP Mass Balance Project represents an effort to collect, verify, analyze, and interpret available data to provide an overall accountability, or site mass balance, for ORGDP RU streams. In addition, data on ORGDP processes and activities and data on Pu, Np, and ^{99}Tc—the primary contaminants of concern in the RU stream—have also been collected, analyzed, and interpreted. Based on this information, the project team has attempted to identify all those activities that (1) created a likelihood of workers coming into contact with concentrated RU constituents through direct physical contact or via airborne dust and/or (2) caused reportable environmental releases of concentrated RU constituents.

The project team analyzed data on receipts, shipments, inventories, product, tails, releases, and other categories—along with available analytical data—in the context of documented historical information on ORGDP processes and activities. Understanding of GDP processes known to concentrate Pu, Np, and ^{99}Tc and of GDP processes and activities known to create potential for exposure to these RU constituents provided additional context for analysis. By correlating mass balance data, analytical data, health physics data, environmental sampling data, and historical information on ORGDP processes, the team was able to identify specific processes, locations, and time periods of importance for potential worker exposure or environmental contamination. These processes, locations, and time periods became the focus of

[1] Following the shutdown of ORGDP in 1987, the facility was known as the Oak Ridge K-25 Site. In 1997, it became the East Tennessee Technology Park (ETTP).

additional assessment to determine the situations that had the potential to create exposure hazards for workers and/or significant environmental contamination.

II. RECYCLED URANIUM AT ORGDP

Receipts

For purposes of this project, RU has been defined as any uranium that has been irradiated in a reactor and as a result contains TRU (e.g., Pu and Np) and fission products (e.g., ^{99}Tc). The methodology applied in this project for identifying ORGDP's involvement with the flow of RU materials involves: (1) the source site and (2) the ^{235}U assay of the material. Sites identified as RU candidate source sites are the U.S. government facilities at Hanford and Savannah River that operated production reactors and used chemical separation processes to extract uranium from irradiated fuel, Harshaw Chemical Company, and foreign customers for U.S enrichment services. Secondary sites providing RU to ORGDP included Paducah Gaseous Diffusion Plant (PGDP), Portsmouth Gaseous Diffusion Plant (PORTS), and the Oak Ridge National Laboratory (ORNL).

ORGDP received a total of 18,654 MTU of RU through three primary pathways:

- **Receipts of 16,268 MT** of RU oxide provided as feed to ORGDP by Hanford, Savannah River, and Harshaw Chemical Company. This oxide was processed in the ORGDP feed production facility.
- **Receipts of 1,294 MT** of RU as UF_6 feed from commercial enrichment customers (primarily nuclear utilities in France, the United Kingdom, and Germany). From 1969 to 1988, 807 MTU was fed to the ORGDP cascade; 486 MTU was shipped to PGDP in 1986; and 1 MTU was returned to France in 1988.
- **Receipts of 1,092 MT** of RU as UF_6 feed from PGDP, ORNL, and PORTS (99.2% from PGDP) during 1953 to 1970. This material was fed into the ORGDP cascade.

Shipments

RU streams exited ORGDP via several pathways:

- Shipment to PGDP and PORTS of RU converted to UF_6 or UF_4.
- Shipment of RU fluorination tower waste ashes to PGDP (which subsequently shipped them to Fernald)
- Shipment of product enriched in the ORGDP cascade to the Y-12 Plant, PORTS, and to private-sector companies fabricating fuel for commercial enrichment customers.
- Shipment of tails from the ORGDP enrichment cascade to PGDP for additional "stripping" in the PDGP cascade.
- Shipment of RU from commercial enrichment customers to PGDP after ORGDP was placed on standby (without re-enriching the RU in the ORGDP enrichment cascade).
- Shipment of cylinder heels at ORGDP to PGDP after ORGDP was placed on standby.

ORGDP shipped a total of 12,141 MT of RU to the following sites:

- PGDP 11,629 MTU
- PORTS 301 MTU
- Y-12 Plant 189 MTU
- ORNL 8 MTU
- Savannah River 11 MTU
- Fernald 2 MTU
- Foreign 1 MTU

Feed and Material Unaccounted For

ORGDP fed 5,915 MT of RU into the ORGDP cascade. Cumulative losses and material unaccounted for (MUF) for RU material at ORGDP totaled 598 MTU. Current inventory at ORGDP is 0. The RU mass balance for ORGDP is summarized in Table ES-1 and Figure ES-1.

Table ES-1. ORGDP RU Mass Balance

Category	MT of RU
ORGDP shipments	12,141
Feed to ORGDP cascade	5,915
Cumulative losses and RU material unaccounted for (MUF)	598
Subtotal	18,654
ORGDP receipts	18,654

III. CONSTITUENTS (Pu, Np & ^{99}Tc) IN RU

The 18,654 MTU of RU received by ORGDP is estimated to have contained the following quantities of the RU constituents of concern:

- Pu: 71.5 g (based on data from RU receipts obtained from correspondence of the ORGDP Laboratory Superintendent). Of this 71.5 g, only 0.01 to 0.04 g is projected to have entered the ORGDP cascade. The overwhelming majority of Pu was concentrated in the ash from the feed plant, and a small fraction was retained as cylinder heels. The ORGDP mass balance for Pu is summarized in Figure ES-2.

- Np: 9 kg (based on ORNL composite sample analysis prior to 1957 and PGDP sample analysis from 1957 to 1967). Of this 9 kg, 0.8 kg is estimated to have entered the ORGDP cascade, along with up to 0.17 kg of Np that was fed to the ORGDP cascade in PGDP enriched product. Approximately 75% of the Np received by ORGDP in RU UO_3 is estimated to have remained in feed plant ash and cylinder heels. Almost 1.5 kg of Np was shipped to PGDP in UF_6 from the ORGDP feed plant. Analysis for Np performed by ORNL in 1955 and early 1957 on composite samples of Hanford and Savannah River RU show much higher concentrations of Np (0.78 ppm Np average) than subsequent analysis reported by Smith (0.24 ppm Np average) for the period from mid-1957 through 1967. This estimate

is based on using the ORNL analysis for estimated Np concentration during 1952 through mid-1957 and the Smith analysis for the period from mid-1957 through 1963, when shipments from Hanford and Savannah River to ORGDP ceased. The ORGDP mass balance for Np is summarized in Figure ES-3.

- ^{99}Tc: 135 kg (based on measurements performed from 1959 to 1973 on Tc content in RU from Hanford and Savannah River). Of this 135 kg, 45 kg is estimated to have entered the ORGDP cascade in the RU feed stream—along with up to 165 kg of ^{99}Tc contained in PGDP enriched product (based on PGDP data for 1972–1982 and ORGDP measurements of ^{99}Tc in PGDP product during 1962–1963). Approximately 70 kg of ^{99}Tc was shipped to PGDP in UF_6 from the ORGDP feed plant. In the ORGDP cascade, ^{99}Tc tended to accumulate at the top of the cascade or to migrate to the purge cascade points at the high end of the plant configuration, where it was trapped and/or vented. The ORGDP mass balance for ^{99}Tc is summarized in Figure ES-4.

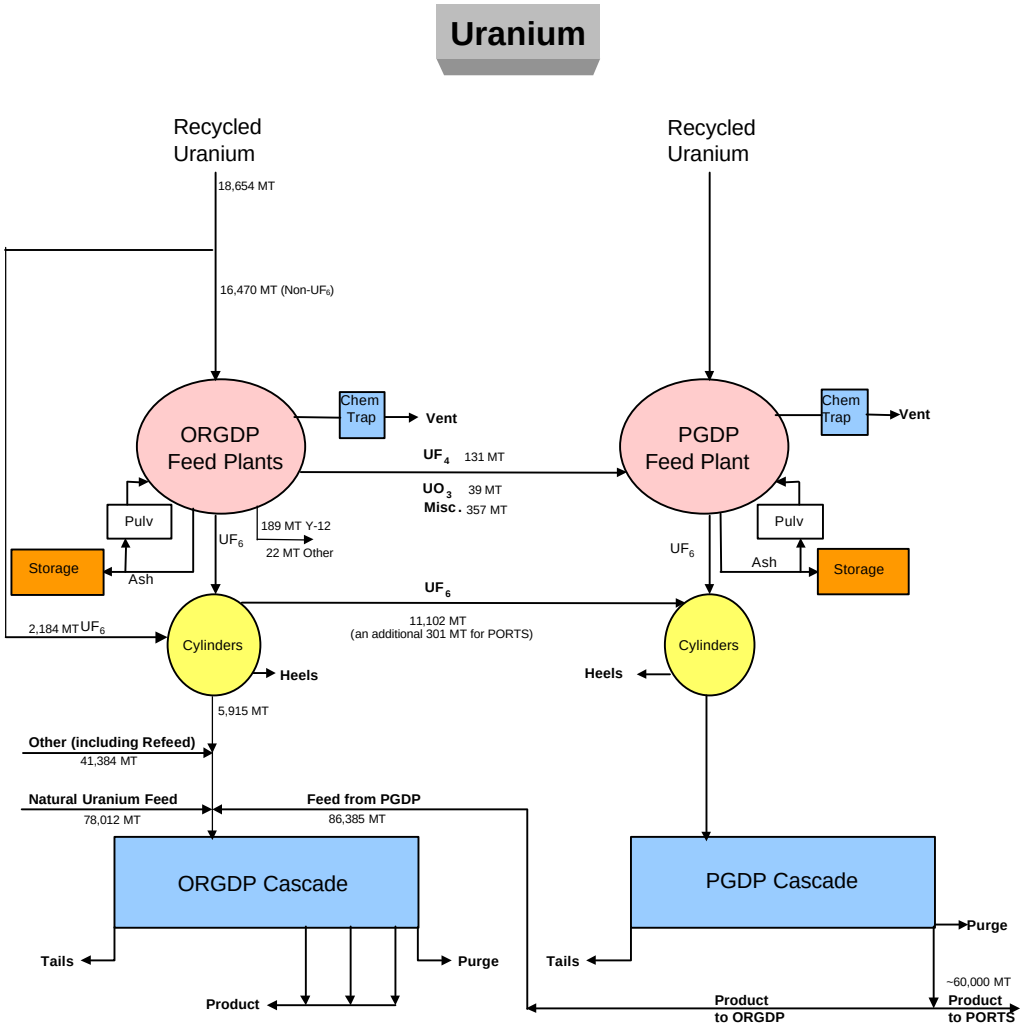
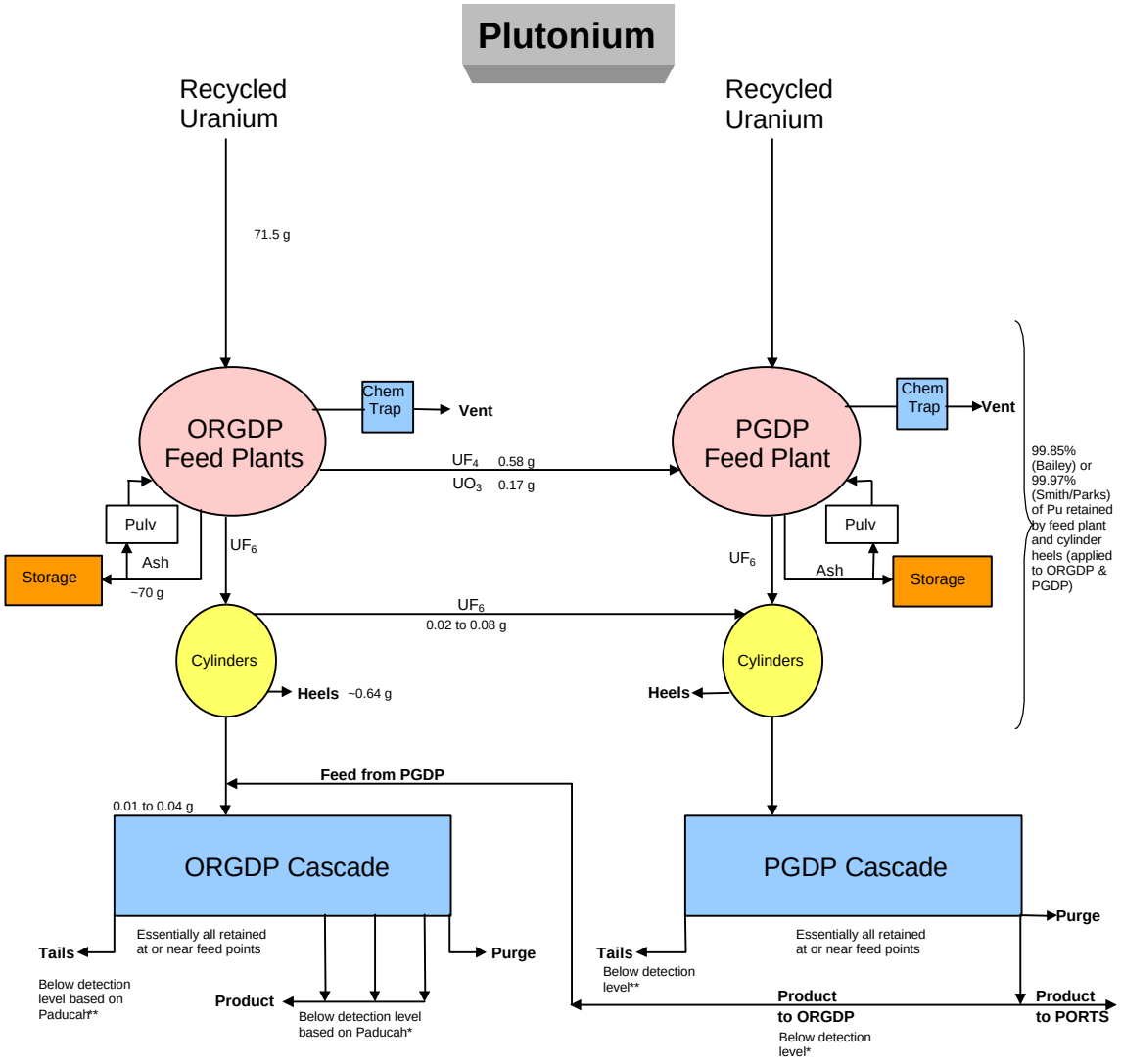


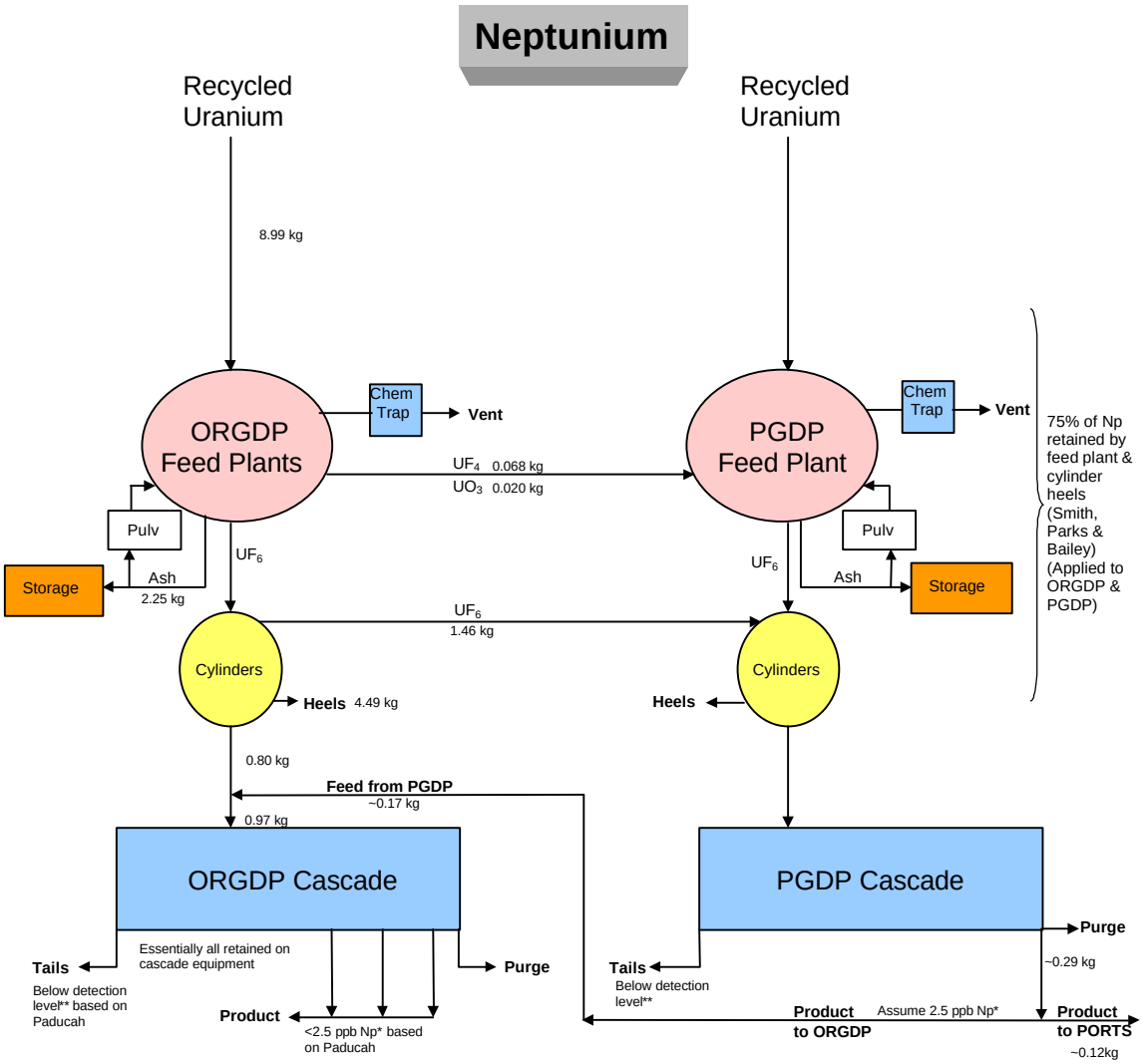
Fig. ES-1. Summary of Recycled Uranium Flow.



**Three product cylinders measured in 1973. Sixty product cylinders measured between 1975 - 1982; all were below detection level of 0.05 ppb initially and 0.01 ppb after 1980 except two cylinders, one showing 0.06 ppb and one showing 0.02 ppb (Smith).*

***Two tails cylinders measured in 1973 <.01 ppb Pu. Routine measurements since 1975 show <0.01 ppb Pu detection level (Smith).*

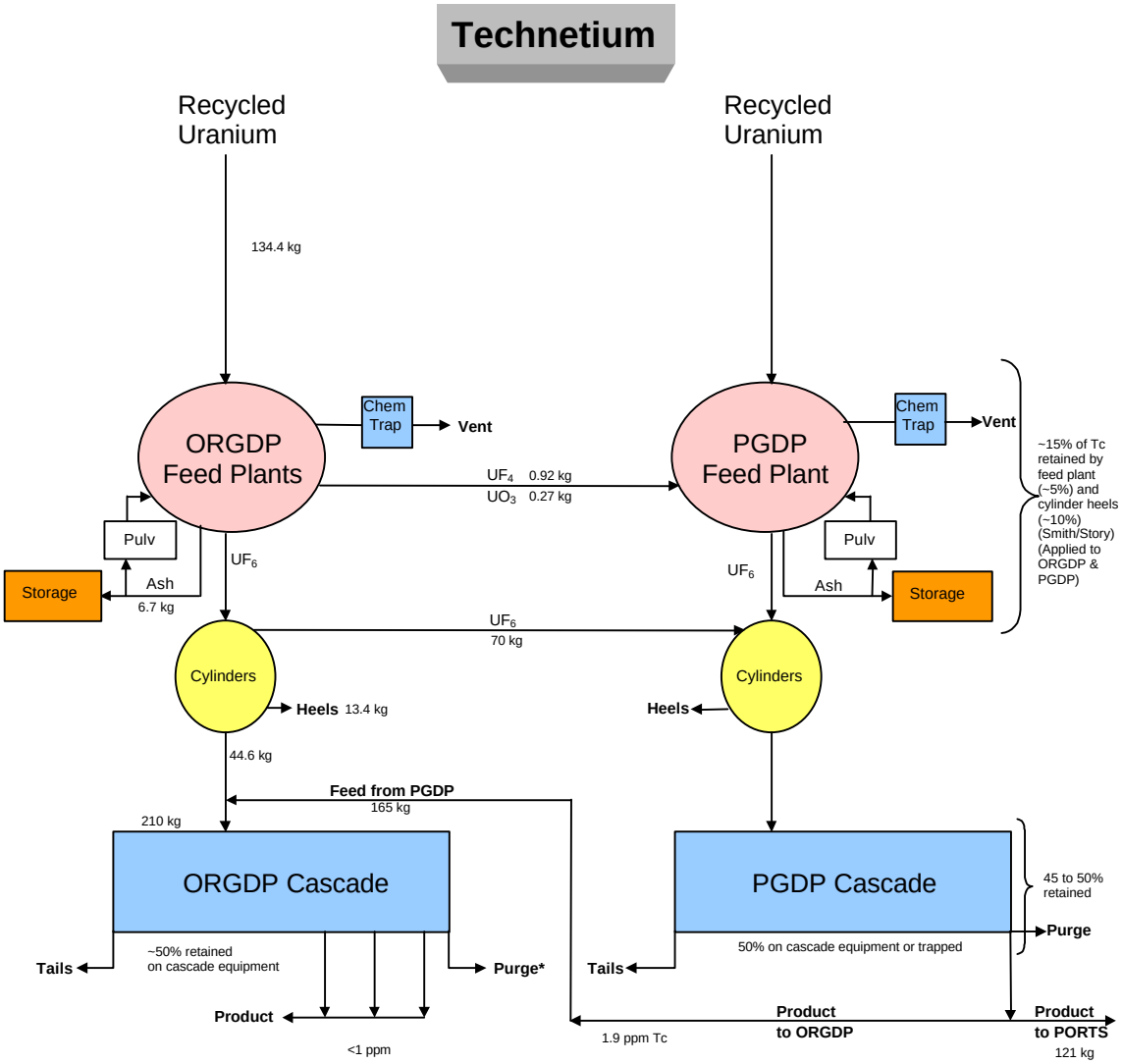
Fig. ES-2. ORGDP Mass Balance for Plutonium.



**60 product cylinders analyzed for Np at Paducah; a few exceeded 5 ppb detection level; highest measurement 27 ppb; most cylinders showed undetectable levels of Np; i.e., <1 and 5 ppb detection levels used (Smith). Assume average Np concentration at Paducah was half of 5 ppb detection level.*

***40 tails cylinders analyzed for Np at Paducah; all were below 1 & 5 ppb detection levels (Smith).*

Fig. ES-3. ORGDP Mass Balance for Neptunium.



**Most of the remaining Tc from the ORGDP cascade (up to ~110 kg) is projected to be in purge cascade equipment, i,e., the K-311-1, K-310-3 system and the K-402-8, K-402-9 system or in the trapped material. A significant quantity of Tc was removed from the purge system by trapping, but the specific quantity is not reported. Very little Tc is expected to have been included in the Product, certainly <1 ppm. Tc in the tails stream is expected to be below detectable levels.*

Fig. ES-4. ORGDP Mass Balance for Technetium.

IV. POTENTIAL FLOW PATHS OF RU WITHIN ORGDP

Once an RU stream entered ORGDP, RU constituents of concern had the potential to reach various facilities and equipment via pathways associated with:

- Oxide conversion to UF_6 for feed
- Cascade buildings and operations
- Uranium recovery operations
- Analytical laboratories

The potential pathways associated with each of these groups of operations are described in the following sections.

Oxide Conversion to UF_6 for Feed Pathways

The process of converting RU oxide to UF_6 for feed for the ORGDP enrichment cascade involved the following potential pathways:

- Unpacking, feeding, and sampling of UO_3
- Collecting ash for uranium recovery and cleaning fluorination tower filters
- Uranium recovery from ash
- Maintenance and repair of fluorination tower and associated equipment

Cascade Buildings and Operations Pathways

ORGDP enrichment cascade operations involved the following potential pathways:

- Feeding UF_6 from cylinders to cascade
- Inadvertent releases of UF_6 within cascade buildings or from piping
- Withdrawal of product
- Withdrawal of tails
- Venting process gas to atmosphere
- CIP/CUP and other equipment removal

Uranium Recovery Operations Pathways

Uranium recovery operations involved the following potential pathways:

- Cleaning heels from UF_6 feed cylinders
- Decontamination of equipment
- Processing of wastes for uranium recovery
- Handling of scrap metal from equipment
- Removal and storage of pond sludge
- Thermal drying and repackaging of pond sludge
- Recovery of uranium deposits from process equipment

- Servicing cascade chemical traps
- Discharge of wastes associated with recovery processes to holding ponds

Analytical Laboratories

Because of the protocols and processes involved in analytical laboratory analysis at ORGDP, these operations created no significant pathways for RU.

V. PROCESSES OR FACILITIES THAT INVOLVED POTENTIAL WORKER EXPOSURE TO RU CONSTITUENTS

Processes and facilities that involved potential worker exposure to RU constituents coordinate closely with the pathways for the flow of RU within ORGDP described in Section IV. Table ES-2 summarizes the activities that were rated by the ORGDP Site Team as "High" in occupational exposure potential—and that consequently have significant implications for potential personnel exposure. For each activity, the table includes information on location, time frame, and RU constituents of concern. (A complete summary of activities at ORGDP with potential for worker exposure to RU is provided in Table 2.4-1.)

Table ES-2. Activities Rated High in Exposure Potential

Location	Activity	Time Frame	Constituents	Occupational Exposure Potential
	1. Oxide Conversion			
K-1131 K-1420	1A. Unpacking, feeding of UO_3 to process, operation and pulling samples * Exposure potential would have been high for brief periods in Jan-Apr 1953 when Pu ranged as high as 40 ppb in material from Hanford	1952-1961 1960-1963	Estimated levels in UO_3 520 ppb Np 4.4 ppb Pu 7,800 ppb Tc 170 ppm ^{236}U	Moderate*
K-1131 K-1420	1B. Collecting ash for uranium recovery and cleaning of tower filters	1952-1961 1960-1963	Estimated levels in ash 13,000 ppb Np 440 ppb Pu 40,000 ppb Tc 170 ppm ^{236}U	High
K-1231 K-1410	1C. U recovery from ash, processes included ash pulverizer	1952-1963 1952-1962	Estimated levels in ash 13,000 ppb Np 440 ppb Pu 40,000 ppb Tc 170 ppm ^{236}U	High
K-1131 K-1410	1D. Maintenance and repair of fluorination tower and associated equipment	1952-1961 1952-1962	Estimated levels 13,000 ppb Np 440 ppb Pu 40,000 ppb Tc 170 ppm ^{236}U	High

VI. PROCESSES OR FACILITIES THAT INVOLVED POTENTIAL ENVIRONMENTAL CONTAMINATION

Process knowledge and a review of documentation narrowed activities that involved potential environmental contamination by the RU constituents of concern to two activities:

- Venting of ^{99}Tc to the atmosphere from the ORGDP enrichment cascade
- Discharges of RU constituents in sludge primarily from the K-1420 decontamination facility to the K-1407-B and –C holding ponds

VI. CONCLUSIONS

Potential Personnel Exposure

The ORGDP Site Team's analysis of ORGDP activities that would have involved potential worker exposure to the RU constituents of concern identified three activities that the team rated "High" in occupational exposure potential and one other activity that was rated high for a brief period over four months in 1953 (Table ES-2). These activities represent the set of ORGDP processes that the Site Team believes involve significant implications for personnel exposure to RU constituents.

With the exception of the unpacking, feeding, and sampling of UO_3 (which only presented "High" occupational exposure potential during a brief period), the occupational exposure potential resulted primarily from hazards posed by fluorination tower ash. An examination of the activities with significant implications indicates that they occurred at the following four locations during the designated time frames:

- K-1131 feed facility (1952–1961)
- K-1231 ash pulverization and uranium recovery facility (1952–1963)
- K-1410 decontamination and uranium recovery facility (1952–1962)
- K-1420 feed facility (1960–1963)

Although both K-1131 and K-1420 performed feed facility functions, K-1131 processed much greater quantities of RU during 1952–1961 than the relatively small portion of K-1420 devoted to feed production did during 1960–1963.

Early in its existence, ORGDP implemented a worker protection program that included worker radiological protection. This program incorporated elements such as personnel protective equipment, personnel monitoring, environmental monitoring, work location surveys, work-time limits on jobs with penetrating radiation, excretion rate limits, periodic examinations of personnel, and Plant Action Level limits. The inhalation of radioactive materials was recognized as the most important source of possible exposure at ORGDP. Consequently, administrative controls were primarily designed to guard against associated hazards.

Worker protection measures in place at ORGDP likely provided substantial mitigation to the risks introduced by the activities rated as "High" in occupational exposure potential. However, dose assessment studies may be warranted as a follow-on activity to provide a more detailed assessment of worker exposure.

Potential Environmental Contamination

An Oak Ridge Dose Reconstruction Project was initiated in 1994 as follow-up to the Oak Ridge Dose Reconstruction Feasibility Study, which recommended a closer examination of past uranium emissions and potential resulting exposures covering the lifetime of the plant. The Task 6 component of the project involved further evaluation of Oak Ridge uranium operations and effluent monitoring records to determine if uranium releases from the ORR likely resulted in off-site doses that warranted further study. The results were documented in the July 1999 Task 6 report entitled *Uranium Releases from the Oak Ridge Reservation—a Review of the Quality of Historical Effluent Monitoring Data and a Screening Evaluation of Potential Off-Site Exposures*. The Task 6 team concluded that earlier estimates of uranium releases had been underestimated. However, based on the decision guidelines from the Oak Ridge Health Agreement Steering Panel, the Task 6 team concluded that while ORGDP uranium releases are candidates for further study, they are not high-priority candidates.

The Task 7 component of the project involved performing qualitative and quantitative screening of various materials of concern at ORGDP and the other DOE Oak Ridge sites. Materials screened included Np and ^{99}Tc. Results were reported in the Task 7 report, *Screening Level Evaluation of Additional Potential Materials of Concern*. Based on the analysis of data, the Task 7 team determined that Np did not warrant further study. Although ^{99}Tc was identified as one of the potential candidates for further study, it was not determined to be a high-priority candidate.

These analyses, along with other information on environmental consequences from ORGDP operations, identify candidate environmental issues for additional study. However, candidate issues related to the processing of RU have not been determined to be high-priority candidates for further study.